

# PATENT SPECIFICATION

1.118.327



NO DRAWINGS

1.118.327

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## COMPLETE SPECIFICATION

### Elastomer Stocks

We, Dow CORNING CORPORATION, of Midland, Michigan, United States of America, a corporation organised under the laws of the State of Michigan, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to elastomer stocks containing an ethylene-propylene terpolymer and has as object to provide such a stock which will yield a cured rubber with improved physical properties, and which is free from the offensive odour that characterises sulphur-vulcanised ethylene-propylene terpolymer rubber.

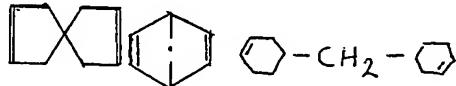
The ethylene-propylene terpolymer is widely known in the art as "E.P.T.". Several varieties of E.P.T. rubber are commercially available and the "E.P.T. rubber" is understood by those skilled in the art to be a specific class of elastomers. The third unit of the terpolymer is a di-unsaturated olefin such as cyclooctadiene-1,5; 1,4-hexadiene; dicyclopentadiene and methylene norbornene.

Detailed information on E.P.T. rubber is available from many sources, including *Rubber Chemistry and Technology*, (1963), pp. 988—999 and 1660 to 1667.

This invention provides an elastomer stock consisting essentially of a mixture of (a) 100 parts by weight of a terpolymer gum of ethylene, propylene, and a di-unsaturated, unconjugated olefin, (b) from 0 to 200 parts of a siliceous filler, (c) from 0 to 10 parts of ZnO, (d) from 2 to 10 parts of an organopolysiloxane containing an average of at least 5  $HR_nSiO_{3-n}$  units per molecule, any other units present being of

the general formula  $(R_mSiO_{3-m})_n$ , where R is a monovalent hydrocarbon radical free from aliphatic unsaturation, n is 1 or 2 and m is 0, 1, 2 or 3, and (e) an amount of platinum or a complex or compound thereof sufficient to catalyse cross-linking of the elastomer stock.

By "di-unsaturated, unconjugated olefin" is meant any unconjugated hydrocarbon diene. Examples of the above are linear dienes such as 2-methylpentadiene-1,4; hexadiene-1,5; hexadiene-1,4; octadiene-1,7; and octadecadiene-1,17; and cyclic dienes such as



and cyclohexadiene-1,4.

The proportions of ethylene, propylene, and unconjugated diene that make up the terpolymers of ingredient (a) is generally not critical, and the formulation of such terpolymers is within the ability of those skilled in the art of organic rubber manufacture. Generally, from 1 to 10 mol per cent of the terpolymer units are unconjugated dienes while the remainder of the units are ethylene and propylene in virtually any proportion.

Any finely-divided or fibrous siliceous filler is suitable for use in this invention, e.g., glass fibres; powdered glass, ceramic; or quartz; or preferably powdered silica. Ingredient (c) is zinc oxide, a known stabiliser for ethylene-propylene rubber, and it is desirable for at least 3 parts by weight of zinc oxide to be present.

5 Ingredient (d) can be any organopolysiloxane of the average unit general formula  $HR_xSiO_{2-x}$ . Examples of R are alkyl and cycloalkyl radicals such as methyl, ethyl,

isopropyl, 2-ethylhexyl, dodecyl, myricyl, and cyclohexyl; and aryl-containing radicals such as phenyl, tolyl, naphthyl, xenyl, and 2-phenylpropyl.

The preferred embodiment of ingredient (d) is a compound of the general formula

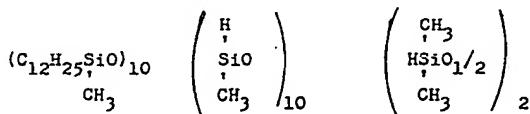
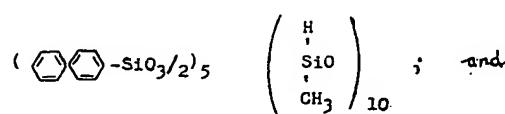
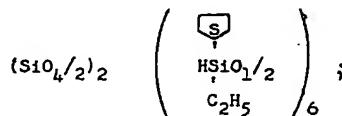
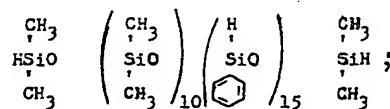


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where x has an average value of from 10 to 90.

Other suitable embodiments of ingredient (d) are



15 Ingredient (d) can contain small amounts of extraneous groups such as silicon-bonded hydroxyl or alkoxy groups. Also, small amounts of other organosilicon compounds can be present in the compositions of this invention, e.g. hexamethyldisiloxane.

20 Ingredient (e), the platinum catalyst, can be present in any known catalytic form, for example, platinum deposited on charcoal or alumina, chloroplatinic acid, platinum complexes with olefins such as cyclohexene, ethylene, propene, butadiene, or styrene, and platinum complexes with organopolysiloxanes which contain aliphatic unsaturation e.g. tetramethyldivinyldisiloxane or phenylallylpolysiloxane.

The amount of platinum present is not critical, but the amount employed generally ranges from 0.001 to 0.01 part by weight, platinum per 100 parts of ingredient (a).

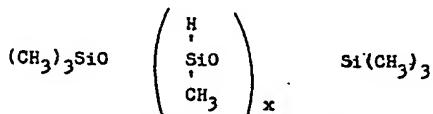
25 The preferred composition of this application is an elastomer stock consisting essentially of (a) 100 parts by weight of a terpolymer of ethylene, propylene, and 1,5-cyclooctadiene; 1,4-hexanediene; a dicyclopentadiene or a methylene norbornene; (b)

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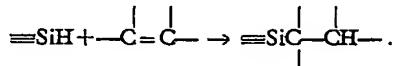
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from 20 to 100 parts of silica filler; (c) from 3 to 7 parts of ZnO; (d) from 2 to 5 parts of a compound of the formula



5 where  $x$  has an average value of from 10 to 90, and (e) from 0.0001 to 0.1 part of a platinum catalyst, calculated on the weight of the platinum.

The curing of the composition of this invention is accomplished through cross-linking of the ethylene-propylene terpolymer by means of ingredient (d), catalysed by platinum in accordance with the well-known reaction:



10 Minor amounts of extraneous materials can be added to the composition of this invention without necessarily rendering them inoperative as curable elastomers. Examples of these are colouring agents, stabilisers, and silicones such as dimethylpolysiloxane.

15 The compositions of this invention cure spontaneously at room temperature at a very slow rate, often sufficiently slow to be inconsequential. Curing can be accelerated to a practical rate by heating the composition at a temperature of about 70° to 150°C, preferably about 100°C. The average time of heating that is required depends upon the curing temperature, and generally runs at the above temperatures from a few minutes to an hour.

20 The cured compositions of this invention have superior physical properties compared with commercial, silica-filled, peroxide-sulphur cured E.P.T. rubber formulations. They are also free from the offensive sulphur odour which is characteristic of sulphur-cured E.P.T. rubber.

25 The following examples illustrate the invention.

EXAMPLE 1  
Two samples of E.P.T. rubber of the following formulations were prepared by milling:

	Sample A	Sample B
	a control sample	
A commercial grade of ethylene-propylene-1-5 cyclooctadiene gum	100 parts by wt.	100 parts by wt.
Powdered silica (~ 5 micron particle size)	80 parts by wt.	80 parts by wt.
powdered ZnO	5 parts by wt.	5 parts by wt.
dicumyl-peroxide	2.7 parts by wt.	—
sulphur	0.32 parts by wt.	—
$(CH_3)_3SiO \begin{array}{c} H \\   \\ SiO \\   \\ CH_3 \end{array} \sim 30_3Si(CH_3)_3$	—	2.5 parts by wt.
platinum (as an activated chloroplatinic acid solution)	—	trace

Both samples were moulded for 10 minutes at 100°C. under steam pressure to yield cured elastomers having the following physical properties:

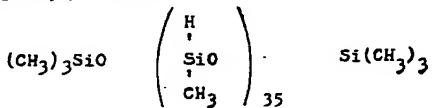
Sample	Durometer	Tensile (lb./in <sup>2</sup> )	Elongation (%)	Tear (die B)
A	58	496	520	81
B	59	738	350	137

EXAMPLE 2

When (a) 100 g. of a terpolymer gum made from 40 mol per cent of propylene, 55 mole per cent of ethylene, and 5 mol per cent of 1,4-pentadiene was milled with (b) 50 g. of glass fibres, (c) 10 g. of a liquid organopolysiloxane having a viscosity of about 2000 cp. at 25°C. and consisting of 30 mol per cent of phenylmethylsiloxane, 23 mol per cent of methylhydrogensiloxane, 1 mole per cent of dimethylhydrogen-siloxane, and 46 mol per cent of phenylsiloxane, and (d) 0.01 g. of platinum complexed with cyclohexene, curing for 15 minutes at 120°C. yielded an elastomer having superior physical properties.

EXAMPLE 3

When (a) 100 parts by weight of a terpolymer gum made from 60 mol per cent of propylene, 37 mol per cent of ethylene, and 3 mol per cent of 1,5-cyclooctadiene was milled with (b) 30 parts of finely-divided silica having a surface area of 400 square metres per gram, (c) 5 parts of ZnO, (d) 3 parts of a compound of the formula



and 0.01 part of the reaction product of chloroplatinic acid and sym-tetramethyl-divinyldisiloxane, curing for one hour at 90°C. yielded an elastomer having superior physical properties.

WHAT WE CLAIM IS:—

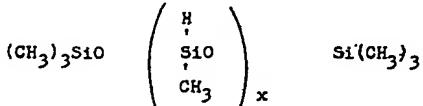
1. An elastomer stock consisting essentially of a mixture of
  - (a) 100 parts by weight of a terpolymer gum of ethylene, propylene, and di-unsaturated, unconjugated olefin,
  - (b) from 0 to 200 parts of a siliceous filler,
  - (c) from 0 to 10 parts of ZnO,
  - (d) from 2 to 10 parts of an organopolysiloxane containing an average of at least 5  $\text{HR}_n\text{SiO}_{\frac{4-n}{2}}$  units per molecule any other units present being of the

general formula  $(\text{R}_m\text{SiO}_{\frac{4-m}{2}})_x$ ,

where R is a monovalent hydrocarbon radical free from aliphatic unsaturation, n is 1 or 2, and m is 0, 1, 2 or 3, and

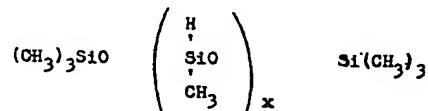
- (e) an amount of platinum or a complex or compound thereof sufficient to catalyse cross-linking of the elastomer stock.

2. An elastomer stock as claimed in claim 1, wherein R is a methyl radical.
3. An elastomer stock as claimed in claim 1, wherein (d) has the general formula



where x has an average value of from 10 to 90.

4. An elastomer stock consisting essentially of
  - (a) 100 parts by weight of a terpolymer of ethylene, propylene, and 1,5-cyclo-octadiene,
  - (b) from 20 to 100 parts of a finely divided silica filler,
  - (c) from 3 to 7 parts of ZnO,
  - (d) from 2 to 5 parts of a compound of the general formula



where  $x$  has an average value of from 10 to 90, and  
 (e) from 0.0001 to 0.1 part of a platinum catalyst for the cross-linking of the  
 elastomer stock, calculated on the weight of the platinum.

5 5. An elastomer stock as claimed in any one of the preceding claims wherein  
 (e) consists of olefin-complexed platinum.

6. An elastomer stock as claimed in any one of the preceding claims 1 to 4  
 wherein (e) consists of a platinum complexed with sym-tetramethyldivinyldisiloxane.

7. An elastomer stock as claimed in any one of the preceding claims 1 to 4  
 wherein (e) consists of chloroplatinic acid.

10 8. An elastomer stock as claimed in claim 1 substantially as described with  
 reference to any one of the Examples.

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